Precision Neutron Inferferometric Search for Evidence of Nuclear Quantum Entanglement in Liquid H_2O-D_2O Mixtures

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(Received 16 September 1998)

A wavelength independent neutron interferometry technique has been applied to measure the scattering length densities Nb of liquid mixtures of H_2O-D_2O near room temperature. The experimental data have been compared to the conventional theory based on two separate liquids, with no deviations greater than 0.4% observed. These results are not consistent with the predicted deviations due to quantum entanglement between protons and deuterons, which are at least an order of magnitude larger than our measured values. [S0031-9007(99)08672-X]

PACS numbers: 67.20.+k, 61.12.Yp, 61.25.Em

Recently, a surprising observation of the anomalous deep inelastic neutron scattering from liquid H_2O-D_2O mixtures was reported [1]. The authors describe this observation as "the first time, direct evidence for short-lived entanglement of adjacent protons (deuterons) in condensed matter at room temperature."

The problem of quantum correlation effects (quantum entanglement—QE) between two particles has been the subject of intensive studies during the past decades (cf. Refs. [6–8] in [1]). It is generally accepted that the time of the existence (decoherence time), if any, of such a correlation (cf. Ref. [7] in [1]) is far beyond the present day instrumental resolution. However, recently it has been suggested [2] that scattering from entangled particles at room temperature may cause a large anomalous scattered field component. This has raised the possibility of experimentally observing QE near room temperature. It has also been suggested that such anomalous scattering can manifest itself as a deviation of the neutron scattering length density [3,4] Nb (N is the atomic density, b is the average neutron scattering length) from the conventional theory. This theory is based upon the assumption that the molecular volumes of the constituents are additive in the mixture. For H_2O-D_2O mixtures deviations as much as 5%–10% [3,4] are predicted from the conventionally expected value $(Nb)_c$ that given by

$$(Nb)_c = \rho_{H_2O}\rho_{D_2O}N_A \times \left[\frac{x_D b_{D_2O} + (1 - x_D)b_{H_2O}}{x_D m_{D_2O}\rho_{H_2O} + (1 - x_D)m_{H_2O}\rho_{D_2O}}\right]. \quad (1)$$

Here ρ are the mass densities of the pure liquids, x_D is the mole fraction of deuterium relative to hydrogen, N_A is the Avogadro number, and m are the molar masses of the molecules. Equation (1) neglects effects due to the excess volumes in H_2O-D_2O mixtures [5], which are negligible on the scale of the present study. Equation (1) is the basis of the widely used technique of contrast variation in neutron scattering, by which the neutron scattering length density Nb of a solvent is matched to that of regions of the dissolved particles. Therefore, the confirmation of the validity of this averaging over isotopes also is of fundamental importance for a vast field of crucial and established experimental research.

Two neutron optical techniques, neutron reflectometry (NR) and neutron interferometry (NI), allow a test of the prediction of QE given in Refs. [3,4]. The use of NI is more preferable in the present case, because it is insensitive to the surface effects that can seriously influence the data of NR experiments. In contrast, the NI technique is sensitive only to the bulk properties of the media. Also, NI is generally a more precise technique for measuring Nb. In Ref. [1] the authors suggest NI and NR experiments to test the predictions of QE. Corresponding experiments were carried out at Hahn-Meitner-Institut (HMI), Berlin, Germany [3–4]. However, the results of these experiments do not provide conclusive evidence for QE.

In this Letter we present the results of a precision NI experiment carried out at the National Institute of Standard and Technology (NIST) in search of evidence of quantum entanglement in liquid H_2O-D_2O mixtures. Unlike the earlier attempt at the HMI [3], the NIST experiment implemented the λ (neutron wavelength) independent nondispersive method [6] in order to avoid potential systematic errors due to the uncertainty in λ. This method has been used to measure the neutron scattering length b of Si with extremely high precision ($\Delta b/b = 0.005\%$) [7]. Such small relative uncertainty is achievable with this method, because it allows for the perfect alignment of the sample surfaces with respect to the crystal planes for the interferometer. When the sample is aligned in this way the phase shift acquired for all the neutron wavelengths is

$$\phi_{nd} = 2dNbt_1, \quad (2)$$

where d is the interferometer lattice plane spacing, and t_1 is the thickness of sample A as shown in Fig. 1.

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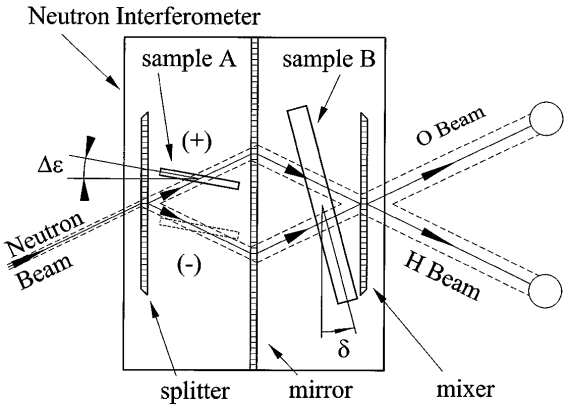

FIG. 1. The experimental arrangement for this experiment.

The experimental layout is shown in Fig. 1. Samples A and B are quartz cuvettes with rectangular cavities that are nominally 0.2 and 3 mm thick, respectively, and contain the same mixture of H_2O-D_2O for any given x_D. Initially, sample A was aligned at an angle ε to be within a few degrees of the exact, nondispersive position ε_0, with $\Delta\varepsilon = \varepsilon - \varepsilon_0$. The experimental data were then collected by rotating the phase shifter (sample B) to an angle δ, and driving sample A between positions $(+)$ and $(-)$. At each position of sample A the neutron intensities in the O and H beams were recorded. An example data set is shown in Fig. 2. This data collection method minimizes the influence of drift of the initial phase of the interferometer itself. The data were then fitted to functions of the standard form,

$$I(\delta, \Delta\varepsilon) = I_0\{1 + C\cos[\Delta\Phi_{\pm}(\delta, \Delta\varepsilon)]\}, \quad (3)$$

where

$$\Delta\Phi_{\pm}(\delta, \Delta\varepsilon) = -2\phi_d \frac{\sin(\theta_B)\sin(\delta)}{\cos^2(\theta_B) - \sin^2(\delta)} + \Delta\phi_{nd,\pm}(\Delta\varepsilon) + \Delta\phi_0, \quad (4)$$

with

$$\phi_d = \lambda N b t_2. \quad (5)$$

Here ϕ_d is the phase shift due to sample B (phase shifter), $\Delta\phi_0$ is the initial phase shift of the interferometer, θ_B is the Bragg angle, t_2 is the thickness of sample B (the phase shifter), I_0 is the incident beam intensity, C is the contrast or fringe visibility, and

$$\Delta\phi_{nd,\pm}(\Delta\varepsilon) = \pm\phi_{nd} \frac{\sin(\theta_B)}{\sin(\theta_B + \Delta_\varepsilon)} \quad (6)$$

is the phase shift due to sample A. This sequence was repeated for various sample angles ε. It has been shown [6] that the difference of the phase shifts when the sample is in $(+)$ and $(-)$ positions depends quadratically on $\Delta\varepsilon$,

$$\Theta(\Delta\varepsilon) = \Delta\Phi_+ - \Delta\Phi_- \approx \phi_{nd}\{2 + (\Delta\varepsilon)^2[1 + 2\cot^2(\theta_B)]\}. \quad (7)$$

The minimum of this parabola corresponds to the perfect nondispersive alignment of the sample surface relative to

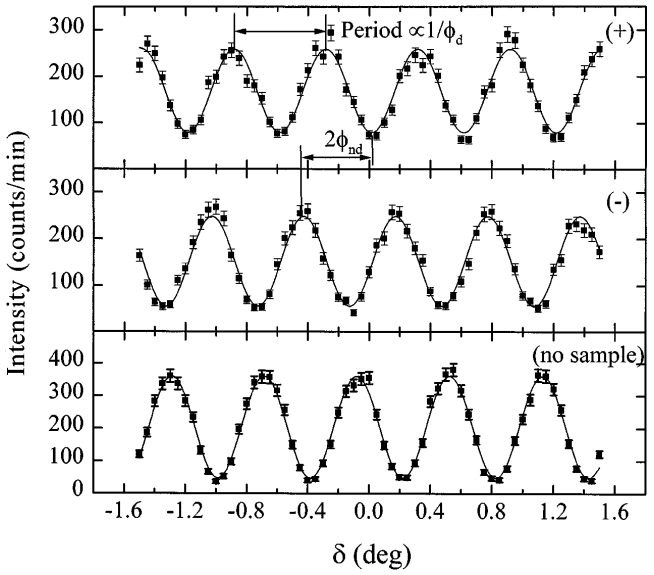

FIG. 2. Examples of neutron interferograms for one of the H_2O-D_2O mixtures. Here $(+)$ and $(-)$ represent the position of sample A shown in Fig. 1. The period of the interferogram is inversely proportional to the total phase shift due to sample B, ϕ_d.

interferometer crystal lattice planes [6]. The experimental data along with a quadratic fit are plotted in Fig. 3, where at angle $\varepsilon_0 = 1.48(2)°$ (minimum of parabola) the sample is nondispersively aligned.

With the sample optimally aligned the phase shifts in positions $(+)$ and $(-)$ are measured repeatedly for mixtures with different x_D. Since replacing the mixtures requires the removal and reinstallation of the cuvettes in the holders, the angular position of sample A is controlled optically with an uncertainty in $\Delta\varepsilon = \pm 0.16°$. This uncertainty in $\Delta\varepsilon$ introduces an error of less than 0.008% in the determination of ϕ_{nd}, which is much less than that required in this experiment. Similar uncertainty is also present in the reinstallation of sample B. However,

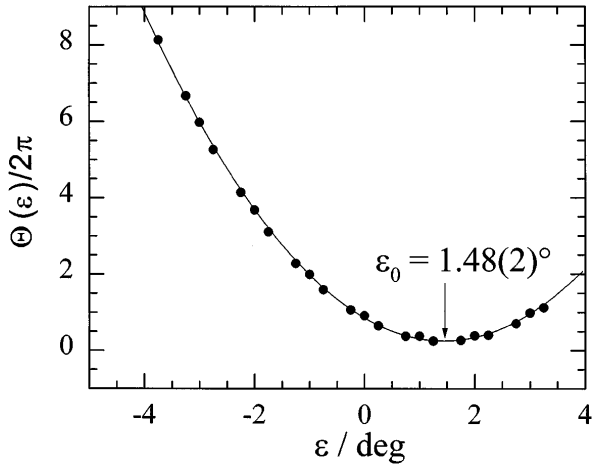

FIG. 3. The difference phase shift as a function of the misalignment angle ε.

the experiment is even less sensitive to such small initial angular misalignment of sample B (phase shifter).

Since the values ϕ_{nd} are found by fitting the data shown in Fig. 2 to Eq. (3), the phase shifts obtained from the fit can be determined only on the interval $[0, 2\pi)$. The following equation then relates the total nondispersive phase shift to the fitted value:

$$\phi_{nd} = \phi'_{nd} \pm 2\pi n\,. \tag{8}$$

The values for n are determined from the total phase shift ϕ_d [Eq. (6)] due to sample B *which are* obtained from fitting the interference data (Fig. 2) to Eq. (3). Then, the total phase shift due to sample A can be calculated by scaling the sample B phase shift ϕ_d by the ratio of their corresponding cuvette cavity thickness (i.e., mixture thickness). Using the values ϕ_d shown in Fig. 4 the values of n were determined by the procedure given by

$$n = \text{Integer part}\left(\frac{2d}{\lambda}\,\frac{t_1}{t_2}\,\frac{\phi_d}{2\pi}\right), \tag{9}$$

where $d = 0.313\,557\,265$ nm, $\lambda = 0.270(1)$ nm, $t_1 = 0.203(1)$ mm, and $t_2 = 0.300(1)$. Although the phase shift ϕ_d is wavelength dependent, the wavelength uncertainty is small enough to allow n to be determined uniquely. The arrangement of using sample B as the phase shifter allowed the determination of the total phase shift of sample B and the phase shift (ϕ'_{nd}) of sample A from the same interference data shown in Fig. 2.

The phase shift differences obtained for the pure H_2O and the pure D_2O samples with respect to the phase shift of the empty cuvette are $(\phi_d)_{H_2O} = -7.27(2) \times 2\pi$ and $(\phi_d)_{D_2O} = 81.54(44) \times 2\pi$. These values are in good agreement with the expected values $-7.22(4) \times 2\pi$ and $81.99(41) \times 2\pi$, respectively, which are calculated from Eqs. (1) and (5) using the known scattering length densities of $(Nb)_{H_2O} = -5.560 \times 10^9$ and $(Nb)_{D_2O} = 6.360 \times 10^{10}$ at 19.0 °C (99.9% isotopic enrichment).

Since [2–4] predict deviations of Nb from the calculated values using Eq. (1), these discrepancies should be present in both phase shift data ϕ_d and ϕ_{nd}. The differences between the experimental values ϕ_d and those calculated from Eqs. (1) and (5) are plotted for the various x_D in Fig. 5. The dashed lines in Fig. 5 represent 1 standard deviation ($\sigma = 0.34$) determined from the scatter of these data. It is seen that this value of σ corresponds to the error bars calculated from the Poisson statistical error of the neutron counts which implies that at this level of precision there is no other source of errors. References [2–4] predict a maximum effect of 5%-10% due to QE in the range $x_D = [0.5, 0.8]$. However, in this range our data show an absence of deviation from that predicted by Eq. (1) no greater than 0.7% at a 68% confidence level.

The values for the nondispersive phase shifts, ϕ_{nd}, are plotted for various x_D in Fig. 6. The difference of these data points from the values calculated from Eqs. (1) and (2) is plotted for each x_D in Fig. 7. The dashed lines in Fig. 7 correspond to the standard deviation $\sigma = 0.031$ evaluated from the scatter of the experimental data. This deviation corresponds to a maximum 0.4% uncertainty with a 68% level of confidence for values of $x_D = [0.5, 0.8]$. The error bars show again the uncertainty contribution from neutron counting statistics alone. Thus the scatter of the data is a measure of the experimental precision determined by the systematics associated with the determination of the phase, such as the variation of temperature and mechanical inaccuracy.

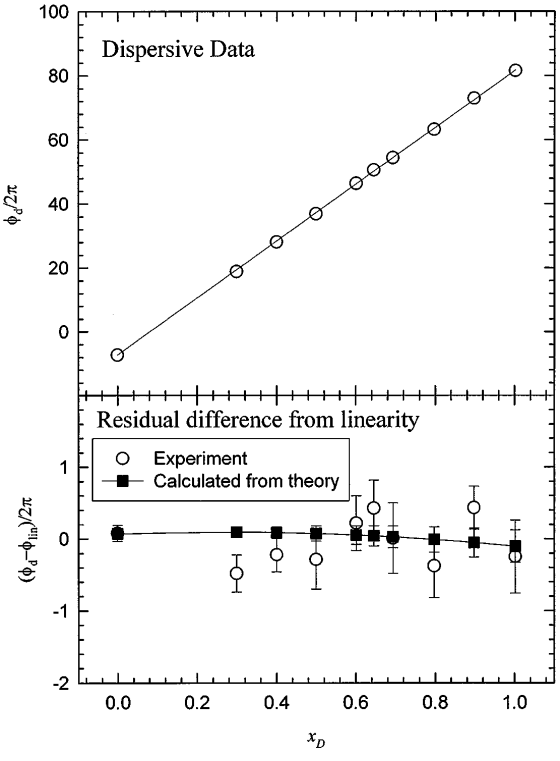

FIG. 4. The top panel is the experimental values for the reduced dispersive phase shifts plotted for various mole fractions x_D. These values have been corrected for the phase shift due to the cuvette which is $35.35 \times 2\pi$. The bottom panel shows the experimental data and the calculated values with the strong linear part subtracted out.

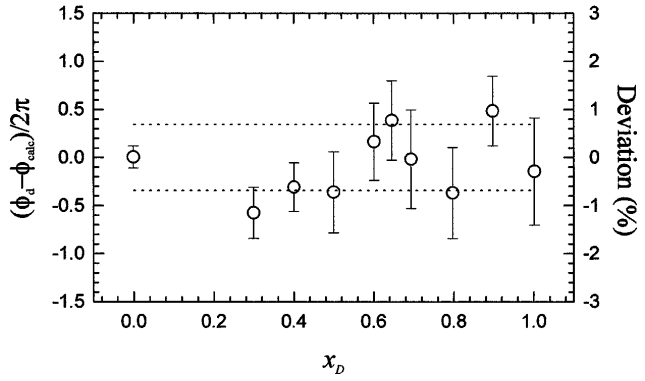

FIG. 5. The residual difference between experiment and theory for $\phi_d/2\pi$. The dashed lines represent the standard deviation $\sigma = 0.34$.

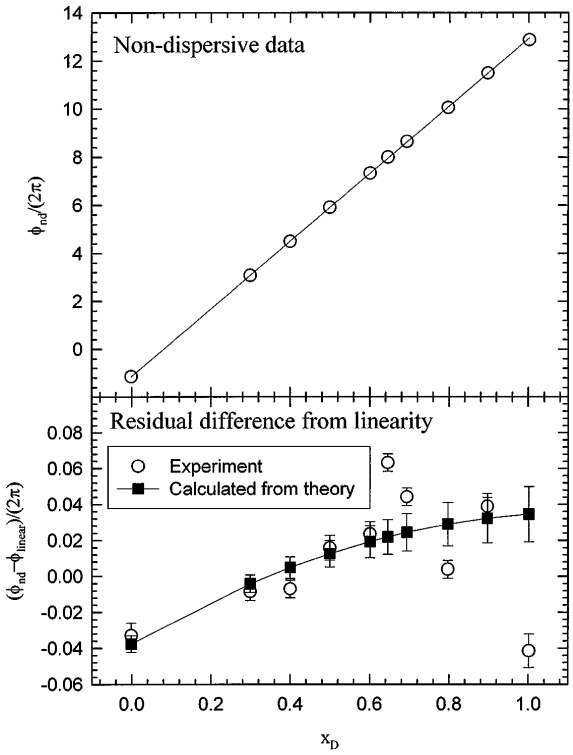

FIG. 6. The top panel is the experimental values for the reduced nondispersive phase shifts plotted for various mole fractions x_D. These values have been corrected for the phase shift due to the cuvette $35.24 \times 2\pi$. The bottom panel shows the experimental data and the calculated values with the strong linear part subtracted out.

We have carried out a NI experiment in order to verify the predictions given in [3,4] which are based on QE in H_2O-D_2O mixtures at room temperature. The use of the nondispersive method enables us to avoid possible systematic errors and to obtain a small relative uncertainty in Nb. We conclude that there is no evidence for deviation greater than 0.4% with a confidence level of 68% from that conventionally calculated on the basis of simple random mixing of two liquids [Eq. (1)]. This result is in disagreement with the predicted deviations in Nb in the range of $5\%-10\%$ [3,4] and confirms the experimental practice of an isotopic contrast variation with high precision. An even more precise experiment is being planned.

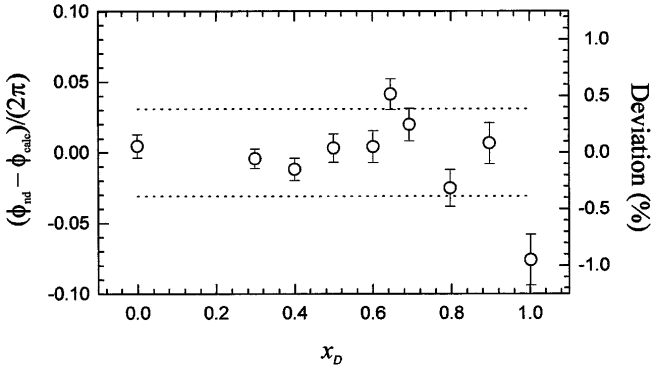

FIG. 7. The residual difference between experiment and theory for ϕ_{nd}. The dashed lines represent the standard deviation $\sigma = 0.031$.

The authors thank Dr. Bert Coursey (Ionizing Radiation Division, NIST) for his helpful discussions on the mass density of H_2O-D_2O mixtures. M. A. also thanks Dr. Terrence Udovic (NIST Center for Neutron Research) for bringing this subject to his attention and suggesting the NIST experiment. A. I. acknowledges support from NSF-Physics Division, Grant No. 9603559 and NIST-UMC Cooperative Agreement No. 8H0008.

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